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ELECTRICAL RESISTIVITY OF THIN EVAPORATED
ALUMINUM FILMS

by



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A THESIS

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The undersigned certify that they have read, and recommend to the Faculty of Graduate Studies and Research, for acceptance, a thesis entitled "ELECTRICAL RESISTIVITY OF THIN EVAPORATED ALUMINUM FILMS", submitted by Choodamani V. SethuRaman in partial fulfillment of the requirements for the degree of Master of Science.

ABSTRACT

The electrical resistivity of evaporated thin polycrystalline aluminum films, ranging in thickness from $50 \text{ \AA}$ - $10,000 \text{ \AA}$ has been measured. It is found that the films differ widely in their structure and cannot be characterised by a single value of ρ_0 and ℓ , ρ_0 being the bulk resistivity which the film would possess if it were made infinitely thick and ℓ the corresponding electron mean free path. The films show a trend towards high ρ_0 and small ℓ with decreasing thickness. The resistivity data have been analysed in terms of Fuchs theory assuming values for $\rho_0 \ell$ obtained from the literature.

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INTRODUCTION

It is well known (Sondheimer, 1952) that the electrical resistivity of a specimen increases above ρ_0 the resistivity of the bulk material, when its dimensions become comparable to or smaller than the electronic mean free path ℓ of the bulk material. Experimental studies of this 'size-effect' in metals are mainly carried out with the purpose of determining the mean free path in bulk metal. Since the product $\rho_0 \ell$ can be expressed in terms of S , the Fermi surface area, or n , the effective density of free electrons taking part in conduction (Ziman, 1964; p.183), the determination of the quantity $\rho_0 \ell$ from size-effect measurements can be used to obtain information about S and n .

The theory of size dependent electrical conduction was first initiated by Thomson (1901) to explain the enhanced electrical resistivity in thin metallic films. The proper derivation was given by Fuchs (1938) for the resistivity of thin films. Fuchs analysis has been applied to the case of thin wires of circular cross section by Dingle (1950) and to wires of square cross section by MacDonald and Sarginson (1950).

Fuchs in his theory assumes that the electron mean free path is isotropic and the Fermi surface spherical. He expresses the increased resistivity ρ in terms of ρ_0 , ℓ and a quantity p , frequently referred to as the specularity parameter, which is the probability that an electron incident on the surface of the conductor is reflected specularly. Because of the assumptions of isotropic bulk mean free path and spherical Fermi surface, Fuchs theory is expected to hold only for monovalent metals in which the electrons occupy only a single band and can be considered to be nearly free.

Though attempts have been made to improve Fuchs theory (these have been reviewed by Larson, 1971), no general theory for metals with non-spherical Fermi surface and anisotropic mean free path has yet been worked out.

As expected, good agreement with Fuchs theory and with the results expected from the free electron model has been obtained by Worden and Danielson (1958) for potassium films, and by Nossek (1955) for a number of alkali metal films. Agreement with Fuchs theory has also been reported for noble metals: by Reynolds and Stillwell (1952) for polycrystalline films of copper and silver; by Kadereit (1967) for polycrystalline gold films; by Larson and Boiko (1964) for single crystal

silver films; and by Chopra and Bobb (1964) for single and polycrystalline gold films. All these data indicate complete diffuse scattering at the boundaries ($p = 0$) in polycrystalline films and partial specular reflection in single crystal films.

The application of Fuchs theory to other metals with valence greater than one and with non-spherical Fermi surfaces is open to question. Though experimental data on a number of polyvalent metals fit Fuchs theory, they yield values of $\rho_0 \ell$ which are considerably greater than the results expected from the free electron model. Resistivity data on aluminum foils by Holwech and Jeppeson (1967) and by Van Zytveld and Bass (1969); on evaporated aluminum films by Bassewitz and Mitchell (1969); on indium foils by Forsvoll and Holwech (1964); and on evaporated indium films by Toxen (1961) all agree with Fuchs theory but give values of $\rho_0 \ell$ which are considerably higher than the value expected from the free electron model assuming three conduction electrons per atom. On the other hand, for both aluminum and indium anomalous skin effect measurements (another size effect in which this time the electron mean free path is compared with the distance to which a high frequency electric field penetrates into the metal (Sondheimer, 1952)) give values of $\rho_0 \ell$ which are close to the theoretical values obtained from the free electron model

(Chambers, 1952; Fawcett, 1960; Dheer, 1961).

The temperature dependence of the resistivity of thin films and wires has also been investigated and deviations from Matthiessen's rule over and above that predicted by Fuchs theory have been observed. This was first observed by Olsen (1958) in indium wires. He found that the increase of resistivity was much greater in the thin than in the thick wires. He proposed that the observed deviations could be attributed to the effect of small angle scattering by phonons at low temperatures. Calculations based on this model have been made by Azbel and Gurzhi (1962) for the temperature dependence of the resistivity of thin films, while Blatt and Satz (1960) and Lüthi and Wyder (1960) have calculated the effect for thin wires. On the other hand from the empirical correlation for indium wires between deviations from Matthiessen's Rule associated with boundary scattering and those associated with dilute impurity scattering Neighbor and Newbower (1969) suggest that the effects of boundary scattering and impurity scattering on the temperature dependence of resistivity may be similar.

In this thesis measurements of resistivity of evaporated polycrystalline aluminum films of thicknesses ranging from $50 \text{ \AA}$ - $10,000 \text{ \AA}$ are reported. The variation of resistivity with temperature from 4.2°K to 76°K has also been investigated for three of these films.

In the first chapter of the thesis Fuchs theory for the resistivity of thin films is presented and its applicability to polyvalent metals is discussed. A brief discussion of conduction in discontinuous films concludes this chapter. In the second chapter the experimental work is described, and in the third and concluding chapter the experimental data are presented and discussed.

CHAPTER I

THEORY

1. Bulk Conduction

According to the modern theory of metals an electron can move without resistance in a perfect lattice. In a metal the resistance to the motion of electrons arises due to the scattering of the electrons (i) by imperfections caused by the thermal motions of ions in the field of which the electrons move, and (ii) by crystalline defects such as vacancies, interstitials, dislocations and the presence of chemical impurities. Assuming that the electron scattering due to the thermal motion and that due to the crystalline defects and impurities is independent, the electrical resistivity $\rho(T)$ can be expressed according to Matthiessen's rule as

$$\rho(T) = \rho_i(T) + \rho_r \quad . \quad (1.1)$$

ρ_r is the residual resistivity due to the scattering of electrons by crystalline defects and impurities and is independent of temperature as long as these lattice defects are not affected by temperature changes. $\rho_i(T)$, the ideal resistivity is strongly dependent in a reversible way on the temperature.

The electrical conductivity of single crystalline metals with cubic symmetry and polycrystalline metals is a scalar and can be written as (Ziman, 1964; p.183)

$$\sigma = \frac{e^2}{12\pi^3\hbar} \int \tau(\vec{k}) |\mathbf{v}(\vec{k})| dS \quad (1.2)$$

where the integral is taken over the Fermi surface and $\mathbf{v}(\vec{k})$ is the velocity of an electron of wave vector $\vec{k}$. $\tau(\vec{k})$, referred to as the relaxation time (which need not be constant over the Fermi surface (Ziman, 1960; p.269)), is the time constant associated with the rate of return to equilibrium of the electron distribution when the external field is switched off. The relaxation time defined in the above sense is a useful concept when applied to scattering of electrons by impurities and also for scattering by lattice vibrations if the temperature is above the Debye temperature (Blatt, 1968; p.121). The electrons taking part in the conduction are treated as quasi-free, with an effective mass m^* which need not be equal to the free electron mass. The mean free path of the electron to which $\tau(\vec{k})$ and $\mathbf{v}(\vec{k})$ refer to is defined as $\ell = \tau(\vec{k}) |\mathbf{v}(\vec{k})|$.

If an average mean free path $\bar{\ell}$ be defined for all the electrons over the Fermi surface then (1.2) can be transformed to

$$\sigma_o = \frac{e^2 \bar{\lambda} S}{12\pi^3 \hbar} = \frac{\bar{\lambda} e^2}{\hbar} \left(\frac{n^2}{3\pi^2} \right)^{1/3} \quad (1.3)$$

where $\bar{\lambda} = \frac{1}{S} \int dS \tau(\vec{k}) |v(\vec{k})|$ and n is the effective free electron density.

2. Fuchs Theory for Conduction in Thin Films

Fuchs analysis for the conduction in thin films is based on the Boltzmann equation for the distribution function of the free electrons. Fuchs assumed an average smooth surface for the film and that all the scattering mechanisms in the bulk metal occur in the film with the only difference that the electrons undergo additional scattering at the boundaries of the thin film. Hence for thin films the total resistivity $\rho_f(T)$ can be written as

$$\rho_f(T) = \rho_i(T) + \rho_r + \rho_t \quad (1.4)$$

where ρ_t is the resistivity due to the scattering of electrons by the surfaces of the film, which will depend on the thickness of the film and the electron mean free path. Hence the above relation implies that (i) there should be a thickness dependent departure from Matthiessen's rule and (ii) the temperature coefficient of resistivity should also become dependent on the thickness.

The Boltzmann equation for conduction phenomena is formed by equating the rate of change of the electron distribution function f due to the external field to the rate of change of f due to the various collision mechanisms, which tend to restore the distribution to its equilibrium value. (The equilibrium distribution is the Fermi-Dirac function).

In general

$$f(\vec{k}+\vec{k}dt, \vec{r}+\vec{r}dt, t+dt) = f(\vec{k}, \vec{r}, t) + \left. \frac{\partial f}{\partial t} \right|_{\text{coll.}} dt. \quad (1.5)$$

In the presence of electrical field E , when the steady state is attained the Boltzmann equation is

$$-e\vec{E} \cdot \frac{1}{\hbar} \nabla_{\vec{k}} f(\vec{k}, \vec{r}) + \vec{v} \nabla_{\vec{r}} f(\vec{k}, \vec{r}) = \left. \frac{\partial f}{\partial t} \right|_{\text{coll.}}. \quad (1.6)$$

In the case of a homogeneous, isothermal bulk metal the distribution function is independent of space coordinates. But in thin films the electron distribution cannot be uniform in space due to the presence of boundaries of the specimen and the boundary conditions at the film surfaces must be imposed on the solution of the Boltzmann equation. These boundary conditions are determined by the nature of scattering at the boundaries of the specimen. Fuchs assumed a boundary condition for the electron distribution function such that a fraction

p of the electrons incident on the surface of the film is reflected specularly and the remainder are diffusely scattered.

Consider a thin film of thickness d lying in the xy plane with the two surfaces being at $z = 0$ and $z = d$. If we assume that all the electrons which reach the surface are scattered diffusely ($p = 0$) then the distribution function for the electrons at $z = 0$ and $z = d$ must be the equilibrium value irrespective of the distribution elsewhere in the film. The distribution function of the electron may be written as

$$f = f_0 + f_1(v, z) \quad (1.7)$$

where f_1 is assumed to be small. When the electric field is applied along the x direction, neglecting the product Ef_1 equation (1.6) reduces to

$$v_z \frac{\partial f_1}{\partial z} - \left. \frac{\partial f}{\partial t} \right|_{\text{coll.}} = \frac{eE}{m^*} \frac{\partial f_0}{\partial v_x} \quad (1.8)$$

Putting in the relaxation time approximation

$$\left. \frac{\partial f}{\partial t} \right|_{\text{coll.}} = - \frac{f - f_0}{\tau} \quad (\tau \text{ is assumed to be isotropic})$$

and applying the boundary conditions that $f(v, z)$ must be zero for electrons at the boundaries of the specimen for completely diffuse scattering ($p = 0$), equation (1.8)

can be solved for $f_1(v,z)$. Assuming a spherical Fermi surface Fuchs obtained for the conductivity σ of the film

$$\frac{\sigma}{\sigma_0} = 1 - \frac{3}{4} \left(\kappa - \frac{\kappa^3}{12} \right) \text{Ei}(-\kappa) - \frac{3}{8\kappa} (1 - e^{-\kappa}) - \left(\frac{5}{8} + \frac{\kappa}{16} - \frac{\kappa^2}{16} \right) e^{-\kappa} \quad (1.9)$$

where $\kappa = d/\ell$, $-\text{Ei}(\kappa) = \int_{\kappa}^{\infty} \frac{e^{-t}}{t} dt$, and

σ_0 and ℓ are the conductivity and mean free path in an infinitely thick film having the same structure and purity as the thin film. The limiting forms of the above expression for $\kappa \gg 1$ and $\kappa \ll 1$ are

$$\frac{\sigma}{\sigma_0} = 1 - \frac{3}{8\kappa} \quad \kappa \gg 1 \quad (1.10)$$

$$\frac{\sigma}{\sigma_0} = \frac{3\kappa}{4} \left[\ln \frac{1}{\kappa} + 0.423 \right] \quad \kappa \ll 1 \quad (1.11)$$

For a general case which assumes that a fraction p of the electrons are scattered specularly at the surfaces Fuchs obtained

$$\frac{\sigma}{\sigma_0} = \left[1 - \frac{3}{2\kappa} (1-p) \int_1^{\infty} \left(\frac{1}{t^3} - \frac{1}{t^5} \right) \frac{1 - e^{-\kappa t}}{1 - p e^{-\kappa t}} dt \right] . \quad (1.12)$$

Lucas (1965) used Fuchs approach to calculate the conductivity of a film with its two surfaces characterised by different specularly parameters p and q .

3. Other Models

According to equation (1.12) if all the electrons undergo specular reflection then the conductivity will be the same as the bulk conductivity. This however applies only to an ideal metal with a spherical Fermi surface and isotropic relaxation time or to a polycrystalline metal with a random angular distribution of the crystallites. The presence of the surfaces in a thin specimen will cause the conductivity of single crystal films with non-spherical Fermi surfaces to be anisotropic even if the bulk conductivity is isotropic (Pippard, 1962). Extension of Fuchs model to a single crystal metal film with a Fermi surface which is an ellipsoid of revolution has been made by Englemann and Sondheimer (1956) assuming diffuse scattering at the boundaries. Price (1960), Parrott (1965, 1966) and Gorkhun and Rashba (1969) have also included specular reflection.

Fuchs model does not take into account the effect of small angle scattering of electrons by phonons at low temperatures. It was pointed out by Olsen (1958) that though these processes contribute negligibly to the resistivity in a bulk metal, they will cause the electrons to strike the surfaces of a film and result in large angle scattering processes when the surface scattering is diffuse. Calculations based on this small

angle scattering mechanism have been made for thin films by Baines (1961), Husstad and Lothe (1967) and by Azbel and Gurzhi (1962).

In all the models mentioned above the boundary conditions proposed by Fuchs are retained. In Fuchs theory the specular parameter p , introduced to describe the scattering of electrons at the boundaries, is treated as though it were independent of the electron wavelength and the angle of incidence. Parrott (1965) and Brandli and Cotti (1965) proposed different boundary conditions. In their formulation the constant specular parameter p is replaced by a step function of the angle of incidence, p being one for angles of incidence greater than a critical angle θ and zero otherwise. Ziman (1960; p.459) has discussed the effect of geometrical roughness on the wavelength dependence of p for normal incidence and his treatment has been extended by Soffer (1967) to include oblique incidence. All these models lead to a saturation of resistivity in the thin film limit but agree with Fuchs theory in the limit of very rough and very smooth surfaces.

4. Application of Fuchs Theory to Polyvalent Metals

In applying Fuchs theory to analyse experimental data the following requirements must be satisfied:

Firstly, the assumptions of spherical Fermi surface and isotropic mean free path over the Fermi surface must be satisfied.

Secondly, the films must be continuous and bounded by plane parallel surfaces.

Thirdly, apart from the difference in thickness, the films should all have the same structure and purity so that they all can be characterised by the same values of ρ_0 , l and also of p .

As already mentioned, in the case of alkali metals and noble metals the application of Fuchs theory is straightforward. In these metals the Fermi surfaces are nearly spherical and the mean free paths can be considered to be isotropic over the Fermi surface. (Though these approximations are less likely to be satisfactory for the noble metals in which the Fermi surface touches the Brillouin zone boundaries (Cracknell 1969, 1971)). Moreover it can be assumed that in these metals one electron per atom contributes to the conductivity so that equation (1.3) can then be used to calculate $\rho_0 l$. For a number of monovalent metals the values of $\rho_0 l$ obtained by fitting experimental data to Fuchs theory have been found to be in good agreement with the theoretically calculated values. This seems to indicate that Fuchs theory can be confidently applied to monovalent metals provided the second, and the third

criterion regarding the uniformity of film structure for all the films is satisfied.

The application of Fuchs theory to polyvalent metals is less straightforward. In these metals the Fermi surface lies in at least two Brillouin zones and hence some distortion in the sphericity of the Fermi surface is to be expected near the zone boundaries. Also there is no reason to believe that the mean free path over the Fermi surface will be isotropic. However the Fermi surface constructed by assuming the free electron model (Harrison, 1959) has been found to be a useful guide in the interpretation of experimental results for a number of polyvalent metals (Cracknell, 1969, 1971).

In the metals aluminum and indium with three valence electrons, the free electron sphere intersects the second, third and fourth zones. Experimental studies of the Fermi surfaces of these metals confirm closely the free electron model so that the area of the distorted Fermi surfaces is not expected to be much different from that of the free electron sphere.

The existing experimental data on these polyvalent metals (Larson, 1971; p.135) show that the value of $\rho_0 \ell$ obtained by applying Fuchs theory to these data are considerably greater than the values calculated from the free electron model. On the other hand,

measurements of anomalous skin effect on these metals show close agreement with the free electron model (Chambers, 1952; Fawcett, 1960; Dheer, 1961).

In all the experimental studies on their specimens it has been assumed that all the samples are identical in structure and purity so that they can be characterised by a single ρ_0 and ℓ . There is always a possibility that this criterion may not be satisfied (Mayadas et al, 1969). If the value of ρ_0 increases with decreasing thickness then it is obvious that the application of Fuchs theory to these data will always yield too large a value of $\rho_0\ell$.

The method described by Cotti (1963) obviates the necessity of preparing samples of different thicknesses with identical bulk resistivities. He has shown that the decay time of eddy currents induced by a magnetic change parallel to the surface of the film defines a resistivity ρ_T which has a functional dependence on ℓ and ρ_0 different from that of the direct current resistivity ρ given by Fuchs. Hence measurement of ρ and ρ_T on the same sample enables one to determine ρ_0 and ℓ . However this method also yielded high values of $\rho_0\ell$ for aluminum and indium (G. Brandli et al, 1964). The Fermi surface area calculated from these values of $\rho_0\ell$ are respectively for aluminum and indium only 43% and 59% of the area expected from the free electron model.

As suggested by Cotti et al (1964), this discrepancy can be resolved if it is assumed that the mean free path varies strongly over the Fermi surface. In such a case the mean free path determined by resistivity size effect measurements cannot be regarded as the mean value defined by (1.3) but will be weighted in favour of those parts of the Fermi surface having the longest free paths. The anisotropy in the resistivity of aluminum films reported by Risnes and Solien (1969) has been explained by them by assuming that the electrons near the Brillouin zone boundaries do not contribute to the current. From their experimental studies of size effect on aluminum and indium Cotti et al (1964) believe that the free paths on the Fermi surface in the third and fourth zones and in the second zone close to the zone boundary must be very short and contribute negligibly to the transport phenomena. Brandli et al (1964) argue that the reason that the measurements of anomalous skin effect lead to values of the Fermi surface close to that accepted on the basis of the free electron model is that work so far has been carried out at frequencies which make the penetration depth shorter than the shortest mean free path on the Fermi surface.

5. Electrical Conduction in Discontinuous Films

In all the above discussions, the films were considered to be continuous so that the mechanism of conduction could be considered to be the same as in the bulk metal.

Depending on the growth stages and on the conditions under which a thin film is formed the structure of the evaporated film may be granular and island-like and may not become continuous until a particular thickness is reached. This thickness at which an evaporated thin film becomes continuous again depends on the metal evaporated, the substrate and other conditions such as the temperature of the substrate, rate of deposition, etc. The conduction mechanism in granular films is entirely different from that in the bulk metal. These films are characterised by high resistivity, many orders of magnitude larger than that of bulk material, and a negative temperature coefficient of resistivity. For these films considerable deviations from ohm's law have been observed at high applied fields. The theory of formation of films and the electrical conduction in the island-like film have been discussed in detail by Neugebauer (1964).

The films reported in this thesis will be considered continuous as none of these characteristics of the island-like film have been observed in our films.

Moreover due to its strong affinity to glass, aluminum is known to form continuous films even at very low thicknesses on glass substrate at normal temperatures (Holland, 1963; p.207).

CHAPTER II

EXPERIMENTAL WORK

1. Preparation of Films

The films were prepared by evaporation in vacuum in an evaporator (a glass bell jar with a diameter of ~12" and 30" in height). The vacuum system consisted of an oil diffusion pump backed by a mechanical pump with a liquid nitrogen trap between the bell jar and the diffusion pump. The trap was always kept cold when the system was evacuated to minimise contamination of the system, especially by hydrocarbons from the pumps. During evaporation liquid nitrogen was passed through a copper tube inside the bell jar.

Fisher-brand 12-550 glass microscope slides were used as substrates. The slides were cleaned with detergent and then slightly fire polished. Leads were soldered to the glass slide with indium and they were also soldered to the copper terminals provided on the substrate holder. During evacuation of the bell jar the substrate was subjected to a glow discharge. This is considered to be effective in removing contaminating films from the substrate and also to accelerate the desorption of gases from the substrate, from the walls and other parts of the vacuum system (Holland 1963;

p.75). Aluminum was used as the glow cathode because of its low sputtering rate. The pressure during the glow discharge was kept constant at 300 microns for about 5 minutes. The system was then pumped down and the glow discharge was allowed to continue till the system was connected to the diffusion pump.

The films were prepared at room temperature by evaporating Al (99.999 % purity) from a tungsten helix, which was resistively heated. The films were defined by a mask on which the substrate could be made to rest. Two sets of films were prepared. The first set which we will call set I, consisted of films in the thickness range $50 \text{ \AA} - 10,000 \text{ \AA}$ were prepared at a deposition rate of $20 \text{ \AA/sec}$, with the substrate 30 cms from the source. The films thicker than $1000 \text{ \AA}$ were prepared with a source to substrate distance of 16 cms and at a rate of $50 \text{ \AA/sec}$. The second set of films, to be referred to as set II hereafter, consisted of films in the thickness range $100 \text{ \AA} - 2000 \text{ \AA}$ and were prepared with a substrate to source distance of 30 cms and an average deposition rate of about $15 \text{ \AA/sec}$.

Prior to evaporation the Al wire was melted and during this period the substrate was shielded from the source by means of a shutter covering the source. In case of set II films the evaporation was commenced

when the ultimate pressure in the bell jar was about 5.0×10^{-7} Torr. During evaporation the pressure rose to about 1×10^{-5} Torr. Set I films were evaporated in better vacuum conditions and the pressure during evaporation did not exceed 3×10^{-6} Torr. Taking into account the geometry of the substrate and the source and following the calculations of Holland (1963; p.141) it was verified that the deposition of Al was uniform throughout the length and breadth of the films. The thickness and the rate of deposition of the films were monitored by means of a quartz crystal monitor.

After the evaporation the specimen was left in the evaporator (under vacuum) for about two hours to allow the films to cool and their resistances to attain steady values. The specimen was then transferred to the cryostat for measurement of its resistance at low temperatures.

To avoid exposure of the films to the laboratory atmosphere and consequent contamination, the bell jar was filled to atmospheric pressure with helium gas. The specimen holder (which was provided with an O-ring) could then be fitted into a brass can. Thus the specimen was always surrounded by helium gas, inside the gas tight brass can. It was found that the resistance of the specimen remained stable for days but

exposing the specimen to air even for short periods resulted in resistance changes.

2. Resistance Measurement

The resistances were measured by the standard four terminal technique. The current passed through the specimen varied from 10 μ A to 5 mA depending on the resistance of the film to be measured. It was verified for a number of films covering the entire range of thicknesses investigated that even for much larger currents no deviation from ohm's law could be observed.

The potential differences were measured with a Guildline Type 9160 photo cell galvanometer amplifier and a type 9461 A galvanometer. With this arrangement it was possible to detect a potential difference of 0.01 μ V. The standard resistors with which the resistances of the films were compared were accurate to at least 0.15 %.

To minimise thermal e.m.f.s connections to the potentiometer circuit were made by means of copper wires as far as possible. Also the usual precaution was taken to measure the potential difference alternately for the standard resistance and film resistance

with normal and reversed currents. For potential selection and current reversal Guildline thermal-free switches were used.

The resistances of the films were first measured at room temperature and then the specimen was transferred to a cryostat for measurement of its resistance at temperatures of about 77.7°K (in liquid nitrogen) and at 4.2°K (in liquid helium). After measuring the resistance of the film at 4.2°K , the resistance was measured again in liquid nitrogen and then at room temperature. No significant change in these resistance values was observed.

The variation of resistance with temperature was measured from 4.2 to 80°K for three of the films (from set II) in a cryostat in which the temperature of the specimen could be maintained steady to within 0.001°K , by means of an electronic controller described by Dauphinee and Woods (1955).

The length and breadth of the specimens were measured with a travelling microscope with an accuracy of better than 0.01 cm .

3. Thickness Measurement

a. Quartz crystal minotor. In employing the quartz crystal for measurement of thickness, use is made of the fact that the change in the resonant

frequency of a quartz crystal due to a small change in the vibrating mass is linearly proportional to this change of mass. Moreover the shift in the frequency is independent of the thickness, density or elastic constants of the material added to the crystal.

To monitor the thickness of the films deposited two $\frac{1}{2}$ " diameter, 5 MHz, AT cut crystals were used. They were mounted identically in the same water cooled copper holder with only one crystal exposed to the source to receive the deposited metal film. The crystals were driven by two oscillators mounted inside the system. The high frequencies from the two were mixed and the change in the difference frequency was used to monitor the thickness of the films deposited. The circuit used was similar to the one described by Vrba (1971).

To estimate the change in the frequency due to the possible rise in the temperature of the crystal during evaporation, the crystal was exposed to the hot bare filament for about 5 minutes. (This is much greater than the time required for evaporation of the films reported in this thesis.) The frequency of the crystal drifted slowly and attained a new steady value. This drift in the frequency was only 6 Hz. The drift in the frequency observed when evaporating thick films at high deposition rates was found to be higher, namely

~ 10-15 Hz. For thinner films the drift was small. To avoid operating in the non-linear range of the crystal, after the crystal was loaded with a mass equivalent to a frequency change of 15,000 Hz the crystal was cleaned and new electrodes (silver was used) were evaporated.

b. Calibration of the Crystal. To know the thickness of the films in terms of the change in the resonant frequency, the quartz crystal was calibrated by measuring the thicknesses of the films (evaporated on optical flats at the position of the substrate) using a Tolansky interferometer (for details see Vrba (1971)). Taking into account the geometry and following the calculations by Holland (1963; p.141) the sensitivity of the crystal for Al was found to be $1.5 \text{ Hz}/\text{\AA}^0$ in agreement with the value given by Lawson (1967).

To calibrate the quartz crystal, when the source was moved closer to the substrate a geometrical correction was applied to the earlier calibration.

CHAPTER III

EXPERIMENTAL RESULTS AND DISCUSSION

1. General Considerations

As already mentioned in Chapter I, one of the difficulties in analysing experimental data on polyvalent metals like aluminum is that the value of $\rho_0 \ell$ is not known. The usual procedure is to fit the data to Fuchs theory to deduce the value of $\rho_0 \ell$ and p . Evaluation of the results obtained depends on the purity and structure of all the samples being equal so that they can be characterised by the same ρ_0 and ℓ . Furthermore if the data on thin films are to give information about the transport parameters in the pure bulk metal then the films must have identical structure to the bulk metal. It is seen from table 1 that our data do not satisfy this criterion. The resistivity of even the thickest film at room temperature (at which the size effects should be negligible) does not approach the resistivity of pure bulk aluminum ($2.65 \mu\Omega\text{.cm}$). Since we have reasons to believe (see sec.2 of this Chapter) that for our films the number of impurities and lattice defects is greater in the thinner films, no attempt has been made to fit our data to Fuchs theory assuming a

single value of ℓ and ρ_0 . Instead we have analysed the resistivity data using the values of $\rho_0\ell$ obtained by other workers.

2. Structure of the Evaporated Films

Though the structure of the films reported in this thesis was not examined, information is available which indicates that the structure of thin films is very sensitive to evaporation conditions. Under the conditions of evaporation of our films they are expected to differ from the bulk metal in several respects.

(i) Since the pressure rose to greater than 10^{-6} Torr during the preparation of the films, this would have resulted in substantial trapping of residual gas molecules and reaction of the films with some of the gases, especially oxidation. Thin films are expected to be more contaminated than the thicker ones.

(ii) Electron microscope studies on evaporated films have shown that these films contain a higher density of lattice defects than the bulk metal or foils which have been prepared by thinning from the pure bulk metal. The possible mechanisms for the formation of lattice defects during the growth of the films have been discussed by Pashley (1964; p.59). There is evidence from the annealing behaviour of films (Belser, 1957; Chopra and Bobb, 1964; Grimes et al, 1970) that there is a greater

density of defects in the thinner films. This lends support to the idea that the evaporated films are initially highly disordered (possibly due to the mechanism of growth in the initial stages and the influence of the substrate) but as the evaporation proceeds there is some improvement in the perfection. Further the atoms of the metal could lose energy due to collisions with the molecules of residual gas near the substrate and in this case their surface mobility is expected to be considerably reduced. Thus the presence of residual gas not only introduces impurities in the evaporated film, but can also stabilise the initial disordered structure of the films by reducing the surface mobility of the metal atoms.

Further evidence for the presence of defects in evaporated films comes from the fact that evaporated films are in a state of large stress and exhibit considerable strength. It is believed that among other factors this high strength is due to the presence of a large concentration of dislocations, other lattice defects and impurities which impede the motion of dislocations (Hoffmann, 1966; p.212).

(iii) Depending on the nature and temperature of the substrate during the film preparation the differential thermal contraction, when the specimen is cooled, can

put the film in a strained condition and can introduce additional defect resistance (Grimes et al, 1970). For example the linear thermal contraction of bulk aluminum relative to its length at 293°K is 415×10^{-5} between 0-10°K and for silica glass it is about -8×10^{-5} at 20°K (Corruccini and Gniewek, 1961).^{*} Hence considerable tensile stress can result in aluminum films evaporated on silica glass at room temperature which are subsequently cooled to 4.2°K for measurement. However the contribution of this thermal stress is expected to be important only when the intrinsic stress (arising due to the imperfections in the film) is small. Thickness dependence of the thermal stresses (generated on cooling from room temperature to liquid-helium temperature) have been observed by Caswell et al (1963) for lead films evaporated on polycrystalline nickel substrate.

(iv) In the evaporated films the grain size is smaller than in well annealed bulk metal and the contribution to the resistivity due to the scattering of electrons at the grain boundaries cannot be neglected (Andrews et al, 1969). The grain size is smaller in impure films as the grain growth is impeded by the presence of impurities. Since thinner films are contaminated more, the grain size is expected to be smaller in thin films than in thicker films.

^{*} This linear thermal contraction is small for all glasses compared to that of aluminum.

From the above discussion, we can infer that under the conditions of evaporation our films are expected to contain a substantial number of imperfections, both impurities and lattice defects, and the density of these imperfections is expected to be larger for thinner films.

3. Analysis of Data

(a) Accuracy of Resistivity Data

The accuracy of resistivity data is determined by the accuracy with which the thickness of the films could be estimated. The quartz crystal monitor was calibrated only for thicker films ($\approx 1500 \text{ \AA}$) which were measured by interferometry with an accuracy of 1 %. The same calibration has been used to estimate the thickness of the thinner films also, which then involves the assumption that the density of the films is independent of the thickness. Wolter (1965) found the density of aluminum films to be constant for thicknesses above $200 \text{ \AA}$; but Hartmann (1965) has reported considerable decrease in density for films thinner than $600 \text{ \AA}$. The accuracy with which the thickness of the films could be determined varied from about 8% for the thinner films to about 1-2% for thicker films ($>1000 \text{ \AA}$), which is the uncertainty introduced due to the drift in the crystal frequency after evaporation is completed.

The resistivity data on set I (evaporated at a pressure of $\approx 3 \times 10^{-6}$ Torr) and set II films (evaporated at a pressure of $\approx 10^{-5}$ Torr) have been presented in tables 1 and 2 respectively.

(b) Temperature Dependence of Resistivity and
Deviations from Matthiessen's Rule

For thin films the deviation from Matthiessen's Rule (to be denoted by DMR hereafter) can be written as

$$\Delta = \rho_{if}(T) - \rho_i(T) \quad (3.1)$$

where $\rho_{if}(T)$, the temperature dependent part of the film resistivity depends on d , the thickness of the film. The DMR Δ will now depend not only on the concentration of imperfections in the film but also on the thickness of the film. The resistivity as a function of temperature measured for the three films (of set II) has been presented in table 3. The values of $\rho_i(T)$ at these temperatures were obtained by interpolation, using the ideal resistivity data on pure aluminum reported by Seth (1969). The calculated Δ have been presented in table 3.

Since the absolute resistivity is accurate to only a few percent for the films investigated, the absolute values of Δ calculated from (3.1) can be in error by as much as 100 % even at high temperatures.

Table 1

Room Temperature Resistivity ρ_{Rm} and Resistivity (at 4.2°K) $\rho_{4.2^{\circ}\text{K}}$ in $\mu\Omega\cdot\text{cm}$ as a function of thickness ($\text{\AA}$) for set I films.

$L/W = 9.06$, where L is the length between the potential contacts and W , the width of the film.

Thickness	ρ_{Rm}	$\rho_{4.2^{\circ}\text{K}}$	Thickness	ρ_{Rm}	$\rho_{4.2^{\circ}\text{K}}$
57	21	17	364	3.96	1.14
78	8.1	5.0	446	3.41	0.798
85	12	9.0	524	3.68	0.822
124	6.07	2.93	615	3.59	0.779
132	6.20	3.07	704	3.17	0.617
158	5.93	2.64	712	3.32	0.759
167	5.24	2.19	945	3.02	0.492
175	4.85	1.88	963	3.20	0.516
219	4.46	1.57	1810	2.86	0.386
223	4.32	1.49	4280	2.77	0.227
299	4.66	1.73	10100	2.83	0.168
320	3.97	1.16			

Table 2

Resistivity (at 4.2°K) $\rho_{4.2^{\circ}\text{K}}$ in $\mu\Omega\cdot\text{cm}$
 as a function of Thickness ($\text{\AA}$) for set II films.

$$L/W = 9.06$$

Thickness	$\rho_{4.2^{\circ}\text{K}}$	Thickness	$\rho_{4.2^{\circ}\text{K}}$
100	6.33	301	2.01
100	8.58	411	1.41
142	6.13	441	1.71
146	3.19	569	1.04
225	2.55	726	0.919
248	1.85	1170	0.651
289	2.97	1770	0.500

Table 3

Ideal Resistivity $\rho_i(T)$ (in $\mu\Omega\cdot\text{cm}$) for aluminum;
 Total film resistivity $\rho_f(T)^*$ and Deviations from
 Matthiessen's Rule Δ in $\mu\Omega\cdot\text{cm}$ (for films of set II)
 as a function of Temperature (in $^{\circ}\text{K}$).

$$L/W = 9.06$$

Film 1 Thickness = 225 $\text{\AA}$

$$\rho_{4.2^{\circ}\text{K}} = 2.557 \mu\Omega\cdot\text{cm}.$$

Film 2 Thickness = 570 $\text{\AA}$

$$\rho_{4.2^{\circ}\text{K}} = 1.041 \mu\Omega\cdot\text{cm}.$$

Film 3 Thickness = 441 $\text{\AA}$

$$\rho_{4.2^{\circ}\text{K}} = 1.706 \mu\Omega\cdot\text{cm}.$$

Temp	$\rho_i(T)$	Film 1		Film 2	
		$\rho_f(T)$	Δ	$\rho_f(T)$	Δ
22.85	0.0013	2.565	0.006	1.047	0.005
26.55	0.0025	2.570	0.010	1.05	0.006
31.12	0.0055	2.580	0.017	1.057	0.01
34.19	0.0086	2.585	0.019	1.063	0.013
40.07	0.0181	2.605	0.03	1.077	0.018
48.78	0.0426	2.645	0.045	1.109	0.024
55.0	0.0695	2.687	0.06	1.131	0.02
62.1	0.1084	2.744	0.08	1.181	0.032
68.1	0.1484	2.802	0.096	1.216	0.027
72.0	0.1773			1.228	0.009
72.4	0.1807	2.837	0.098		
77.8	0.2254	2.881	0.098		

Table 3 (continued)

Temp.	ρ_i (T)	Film 3	
		ρ_f (T)	Δ
23.05	0.0014	1.711	0.004
24.25	0.0017	1.712	0.005
27.27	0.0029	1.716	0.008
28.18	0.0034	1.718	0.009
32.0	0.0063	1.725	0.013
37.75	0.0138	1.736	0.016
41.32	0.0208	1.745	0.018
43.70	0.0266	1.760	0.028
48.90	0.0431	1.780	0.031
56.55	0.0772	1.819	0.037
62.05	0.1081	1.851	0.037
65.40	0.1296	1.871	0.036
69.30	0.1571	1.896	0.033
72.90	0.1847	1.925	0.035
75.00	0.2017	1.955	0.047

* Although the uncertainty in determining the cross section of the films varied from about 2% for thick films to about 10% for thin films, in this table the resistivity values have been given to four figures as the uncertainty in the measurement of relative resistances is less than 0.1%.

4. Discussion of Results

(a) Thickness Dependence of the Resistivity

The resistivity data have been plotted in Fig. 1, Fig. 2 and Fig. 3 along with the curves obtained from Fuchs theory (from eqn. 1.9). The values of $\rho_0 l$ used to generate these curves are respectively $11.5 \times 10^{-12} \Omega \cdot \text{cm}^2$ (obtained by Bessewitz and Mitchell (1969) from size effect measurements on evaporated aluminum films of thicknesses varying from 2000 $\text{\AA}$ to 36,000 $\text{\AA}$), $8.2 \times 10^{-12} \Omega \cdot \text{cm}^2$ (obtained by Hollwech and Jeppeson (1967) from resistivity data on pure aluminum foils with thicknesses between 0.017 and 0.95 mm.), and $4.9 \times 10^{-12} \Omega \cdot \text{cm}^2$ (from anomalous skin effect measurements, Chambers (1952)). Though evidence for partial specular reflection in thin polycrystalline gold films has been obtained by several authors (for references see Larson (1971)), and in thin polycrystalline silver films by Estermann and Schlesinger (1970), most of the experimental data on polycrystalline metal films indicate that the surface scattering is diffuse. We have therefore assumed $p = 0$ for our films.

It is seen from Fig. 1 that comparison of the experimental data with Fuchs theory yields for the mean free path a value of $\approx 10,000 \text{\AA}$ (for films $> 200 \text{\AA}$) at 4.2°K. This value of mean free path at 4.2°K appears

Fig. 1

Thickness Dependence of the Resistivity of the films.

————— Resistivity as a function of thickness according to Fuchs theory, with ℓ as parameter.

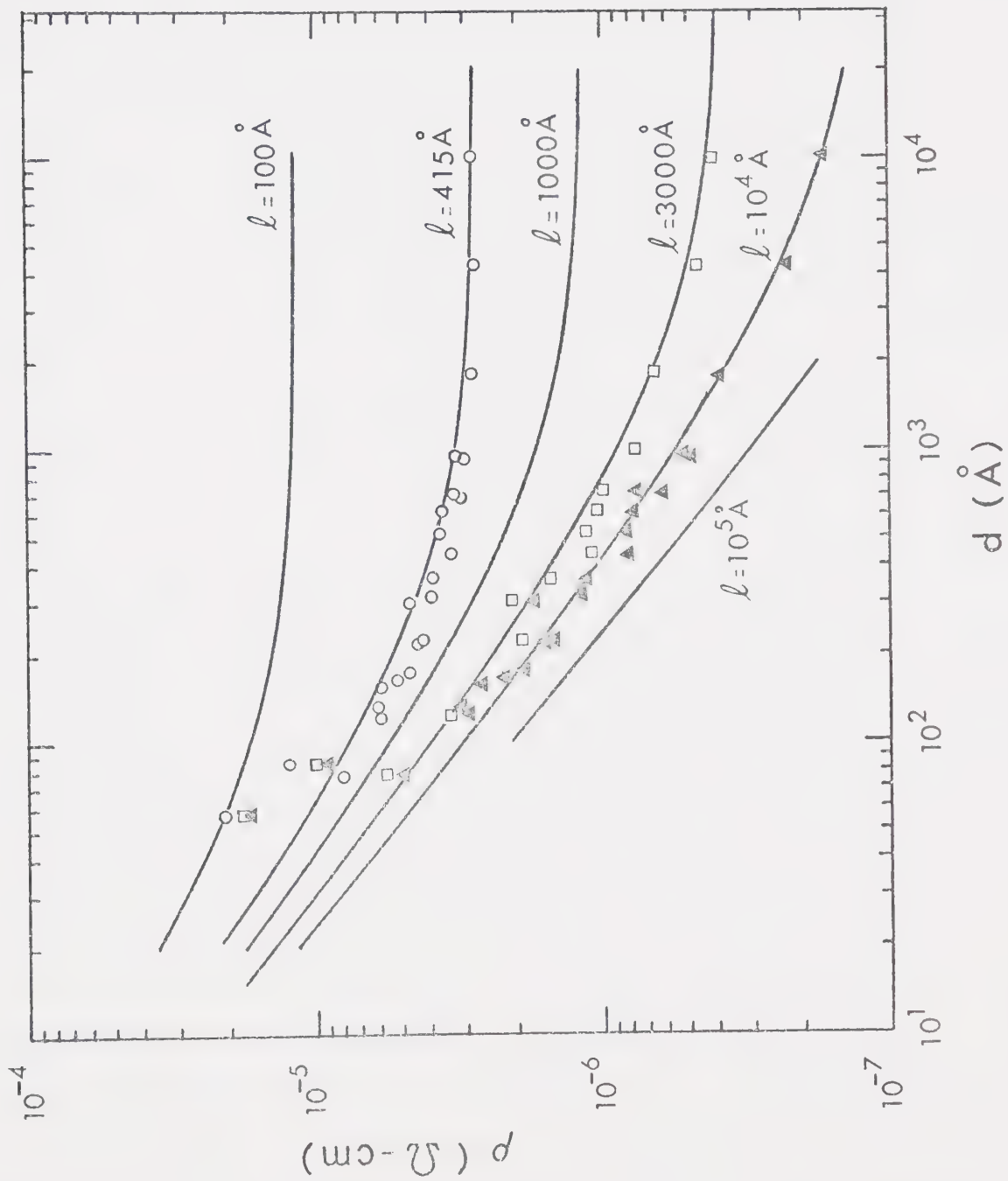
The value of $\rho_0 \ell$ is assumed to be $11.5 \times 10^{-12} \Omega \cdot \text{cm}^2$.

▲, O, □ Represent the experimental data on set I films (evaporated at a pressure of $\approx 3 \times 10^{-6}$ Torr).

▲ Resistivity of the films at 4.2°K.

O Resistivity of the films at room temperature.

□ Resistivity of the films at 77.8°K.



to be reasonable. But the value of ℓ at room temperature is about $415 \text{ \AA}$ for thicker films and seems to be even greater for thinner films. This then implies that thinner films are more perfect than the thicker ones. On the contrary, in view of the factors discussed in sec. 2 of this chapter deterioration in film quality with decreasing film thickness is to be expected. For this reason it appears that our films cannot be characterised by this value of $\rho_0 \ell$ both at room temperature and at 4.2°K .

Considering Fig. 2, the value of $8.2 \times 10^{-12} \text{ } \Omega.\text{cm}^2$ for $\rho_0 \ell$ gives a mean free path of about $300 \text{ \AA}$ at room temperature. At 4.2°K the mean free path for the two thickest films is about $6000 \text{ \AA}$ but decreases with decreasing thickness. The mean free path calculated as a function of thickness for this value of $\rho_0 \ell$ has been plotted in Fig. 4. It is seen that the resistivity data at room temperature (for films $>150 \text{ \AA}$) appear to be in fair agreement with the curve for $\ell = 300 \text{ \AA}$. This value of ℓ at room temperature is greater (about twice) than the average mean free path determined from equation (1.3) assuming three conduction electrons per atom contribute to the conductivity. It is evident from the discussion in section 2 of this chapter that there are other thickness dependent factors which can contribute

Fig. 2

Thickness Dependence of the Resistivity of the films.

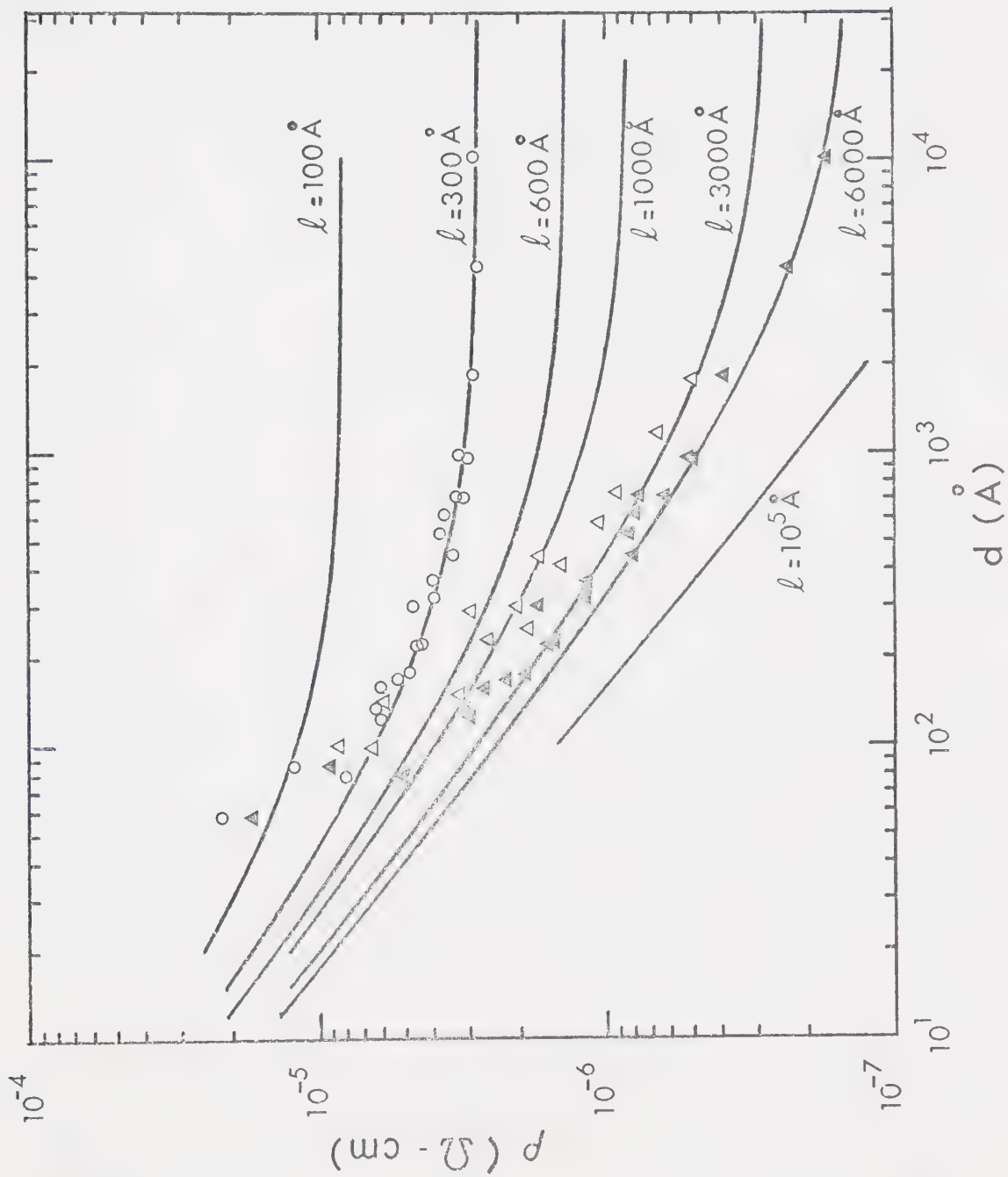
———— Resistivity as a function of thickness from
Fuchs theory with ℓ as parameter. $\rho_0 \ell =$
 $8.2 \times 10^{-12} \Omega \cdot \text{cm}^2$.

▲, O Represent the experimental data on set I films.

▲ Resistivity of the films at 4.2°K .

O Resistivity at room temperature.

Δ Resistivity of the set II films at 4.2°K .



to the observed increase in the resistivity with decreasing thickness. Hence the agreement of the resistivity data at room temperature with Fuchs curve for $\ell = 300 \text{ \AA}$ cannot be considered to be brought about by the true size effect alone (which arises due to the pure geometrical limitation of mean free path by boundary scattering), but also must be due to the systematic increase in ρ_0 with decreasing thickness. This implies that the mean free path will also vary with the thickness decreasing with decreasing thickness of the film. Consequently the value of $\rho_0 \ell$ for our films must also be less than $8.2 \times 10^{-12} \text{ } \Omega \cdot \text{cm}^2$.

As already mentioned in section 4 of Chapter I, the large value of $\rho_0 \ell$ (which is related to the area of the Fermi surface) and hence large ℓ determined from their size effect measurements on indium and aluminum plates were explained by Cotti et al (1964) to be due to the anisotropy of the mean free path. They assumed that the mean free path in those regions of the Fermi surface near the zone boundaries to be very short compared to the mean free path in large segments (those parts of Fermi surface in second zone) which are not close to the zone boundaries. At low temperatures the anisotropy is further enhanced by the electron-phonon umklapp scattering near the zone boundaries. Cotti et al (1964) and Hollwech

and Jeppeson (1967) carried out size effect measurements at low temperatures on annealed samples with purity comparable to that of the pure bulk metal. But for our films which have considerable imperfections the anisotropy of the mean free path should be less serious at low temperatures in the residual resistivity region and may be considerably less at room temperatures.

Finally in Fig. 3 we have plotted our data on set I films along with the curves from Fuchs theory assuming $\rho_0 \ell = 4.9 \times 10^{-12} \Omega \cdot \text{cm}^2$. In this case at room temperature we obtain for the mean free path values which vary from 180 Å to 30 Å (for the thinnest film). At 4.2°K ℓ decreases from 3000 Å for the thickest film to 40 Å for the thinnest film (Fig. 5). These low values of the mean free path appear to be reasonable considering the fact that our films are expected to have considerable impurities and defects and are characterised by low resistivity ratios. As already mentioned, since at room temperature the anisotropy of the electron-phonon scattering (which is the dominant mechanism for restoring electron distribution to equilibrium) should not be large, the mean free path determined by this value of $\rho_0 \ell$ (which is near the value $4 \times 10^{-12} \Omega \cdot \text{cm}^2$ obtained by assuming three free electrons per atom) may be the most reasonable estimate for the average mean free path at room temperature

Fig. 3

Thickness Dependence of the Resistivity of the films.

———— Resistivity as a function of thickness from
Fuchs theory with ℓ as parameter. $\rho_0 \ell =$
 $4.9 \times 10^{-12} \Omega \cdot \text{cm}^2$.

▲, O Represent experimental data on set I films.

▲ Resistivity of the films at 4.2°K .

O Resistivity at room temperature.

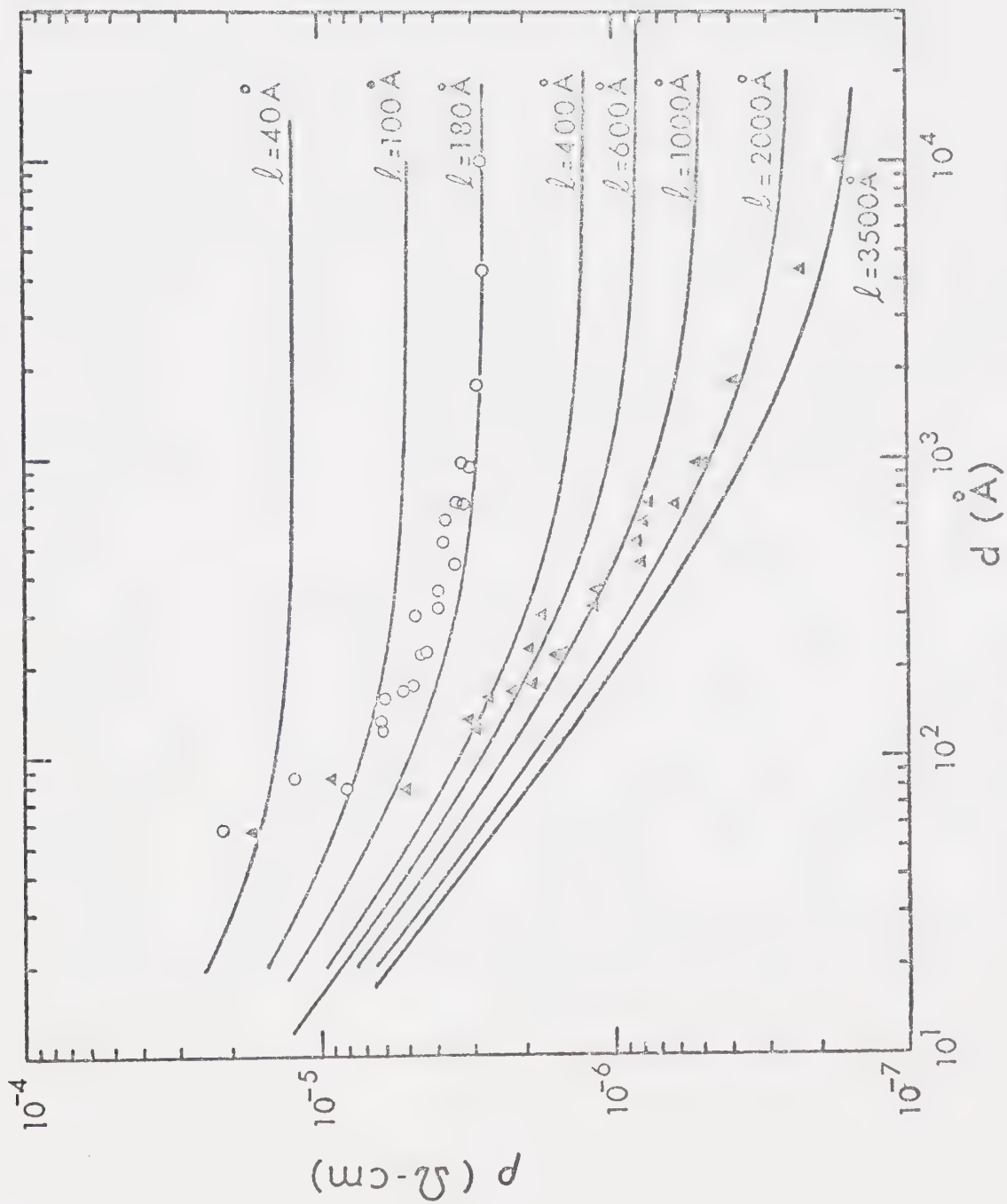


Fig. 4 and Fig. 5

Calculated Mean Free Paths (at 4.2°K)(in set I films) as a function of thickness.

The mean free paths have been calculated according to Fuchs theory from the resistivity data (at 4.2°K) assuming $\rho_0 l = 8.2 \times 10^{-12} \Omega \cdot \text{cm}^2$ (Fig. 4) and $\rho_0 l = 4.9 \times 10^{-12} \Omega \cdot \text{cm}^2$ (Fig. 5).

In Fig.4 ● represent the mean free paths estimated from Fig.2.

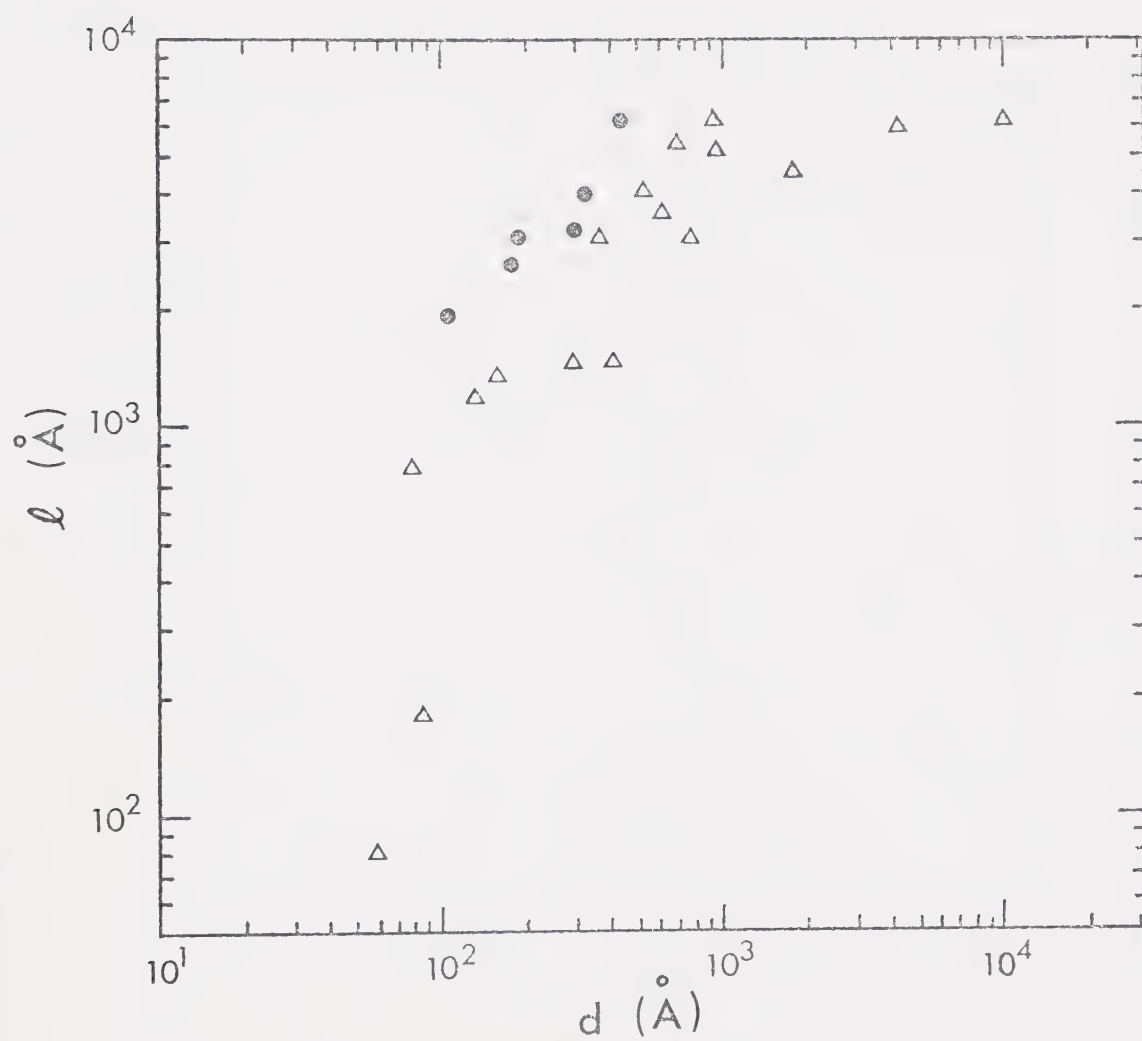


Fig.4

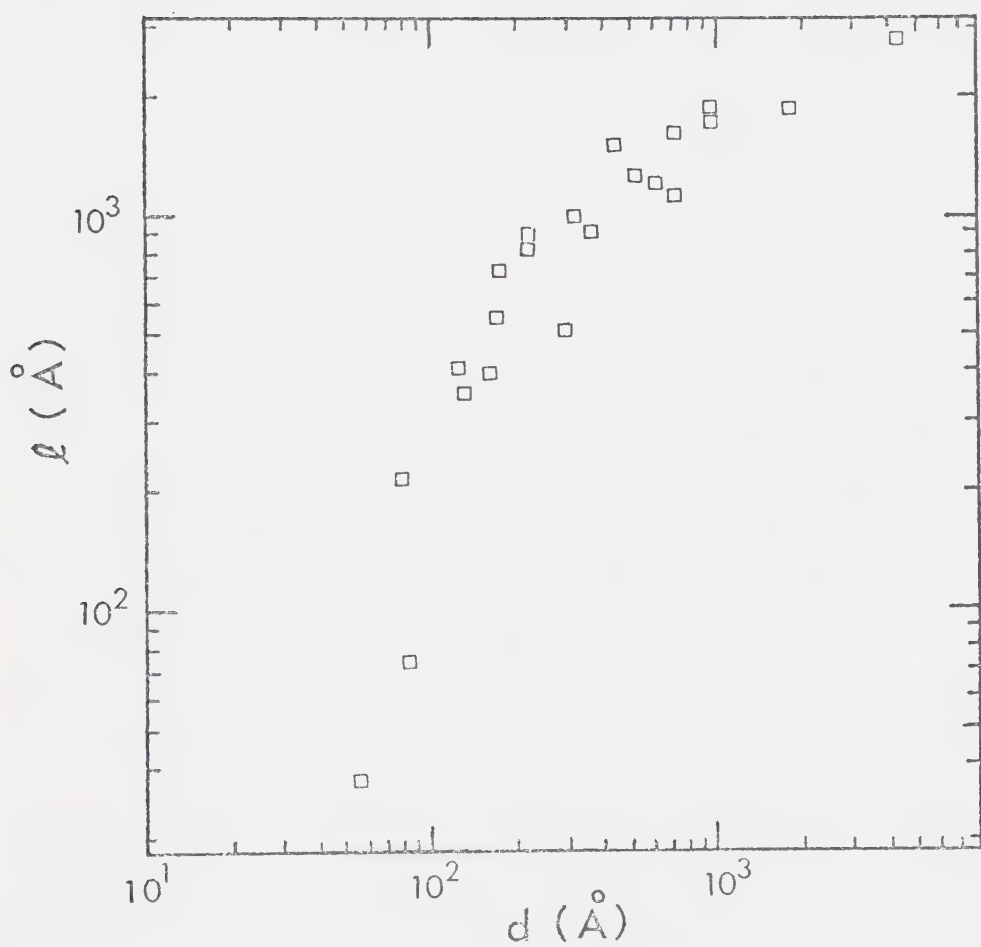


Fig. 5

for our films. At low temperatures however it is not clear what value of $\rho_0 \ell$ should be used to describe our data as the degree of anisotropy may be different for impurity scattering which dominates at low temperatures.

(b) Temperature Dependence of Resistivity

This was investigated for three films from set II. The resistivity of these films as a function of thickness (table 2) is plotted in Fig.2. Comparing the resistivity data of the two sets of films it is seen that the resistivity of set II films is high compared to those of set I films. This difference seems to be higher for thinner films. This indicates that the quality of the films evaporated in a poor vacuum deteriorates more with decreasing thickness than it does for films evaporated in better vacuum conditions.

The DMR has been calculated from Fuchs theory for the three films assuming the values of ρ_0 and ℓ obtained from Fig.2 (i.e. assuming $\rho_0 \ell = 8.2 \times 10^{-12} \Omega \cdot \text{cm}^2$). The calculation of resistivity as a function of temperature from Fuchs theory involves calculating the mean free path ℓ at different temperatures. This is done by assuming (i) the value of $\rho_0 \ell$ to be temperature independent and (ii) that the temperature dependent part of ρ_0 varies in a manner identical to the ideal resistivity of

Fig.6, Fig.7, and Fig.8

Temperature Dependence of Deviations from Matthiessen's Rule.

Fig.6

Film Thickness = $225 \text{ \AA}^{\circ}$.

— DMR Δ according to Fuchs theory for $\ell = 850 \text{ \AA}^{\circ}$, $\rho_0 \ell = 8.2 \times 10^{-12} \Omega \cdot \text{cm}^2$.

o Observed Δ .

Fig.7

Film Thickness = $570 \text{ \AA}^{\circ}$.

— DMR Δ from Fuchs theory for $\ell = 2000 \text{ \AA}^{\circ}$, $\rho_0 \ell = 8.2 \times 10^{-12} \Omega \cdot \text{cm}^2$.

□ Observed Δ .

Fig.8

Film Thickness = $441 \text{ \AA}^{\circ}$.

— DMR Δ from Fuchs theory for $\ell = 960 \text{ \AA}^{\circ}$, $\rho_0 \ell = 8.2 \times 10^{-12} \Omega \cdot \text{cm}^2$.

Δ Observed Δ .

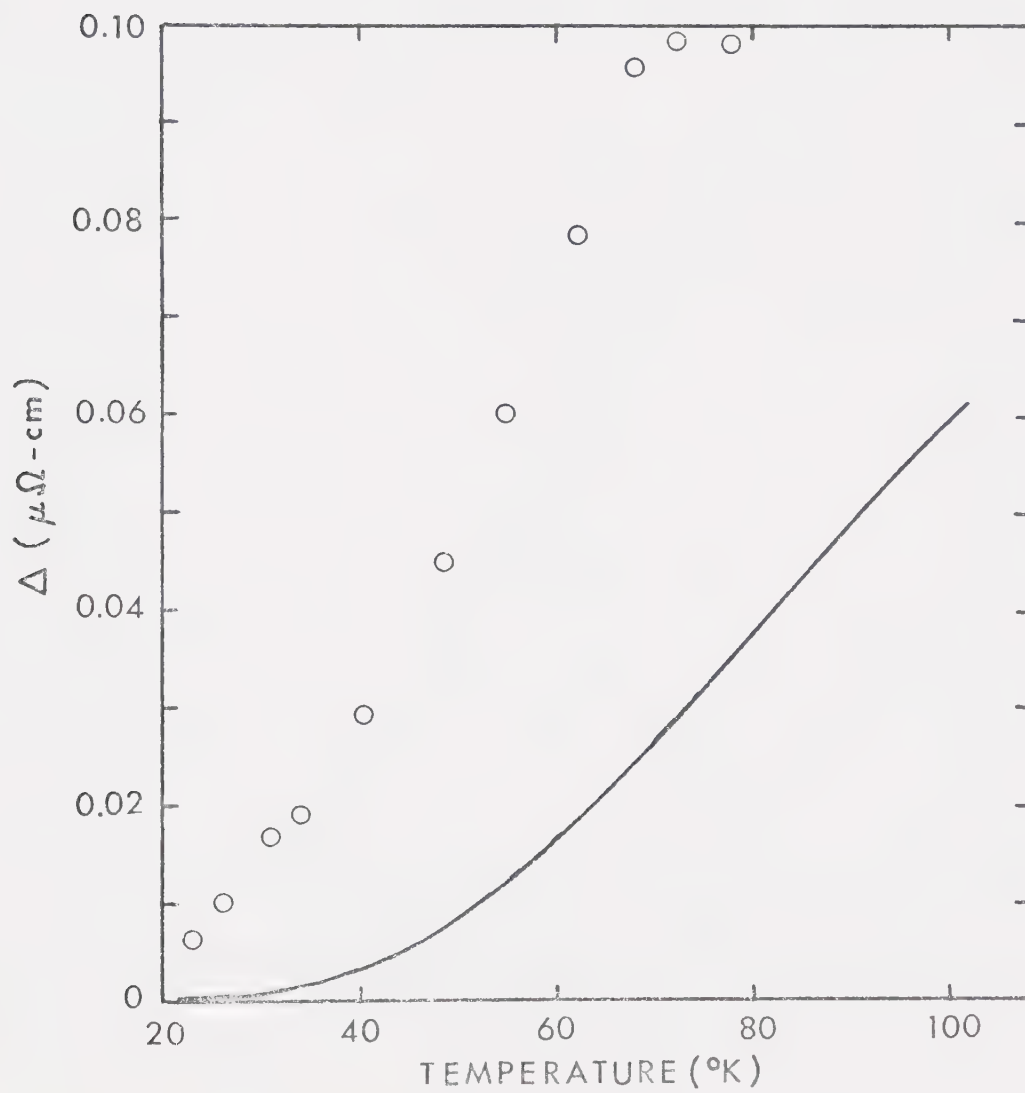


Fig.6

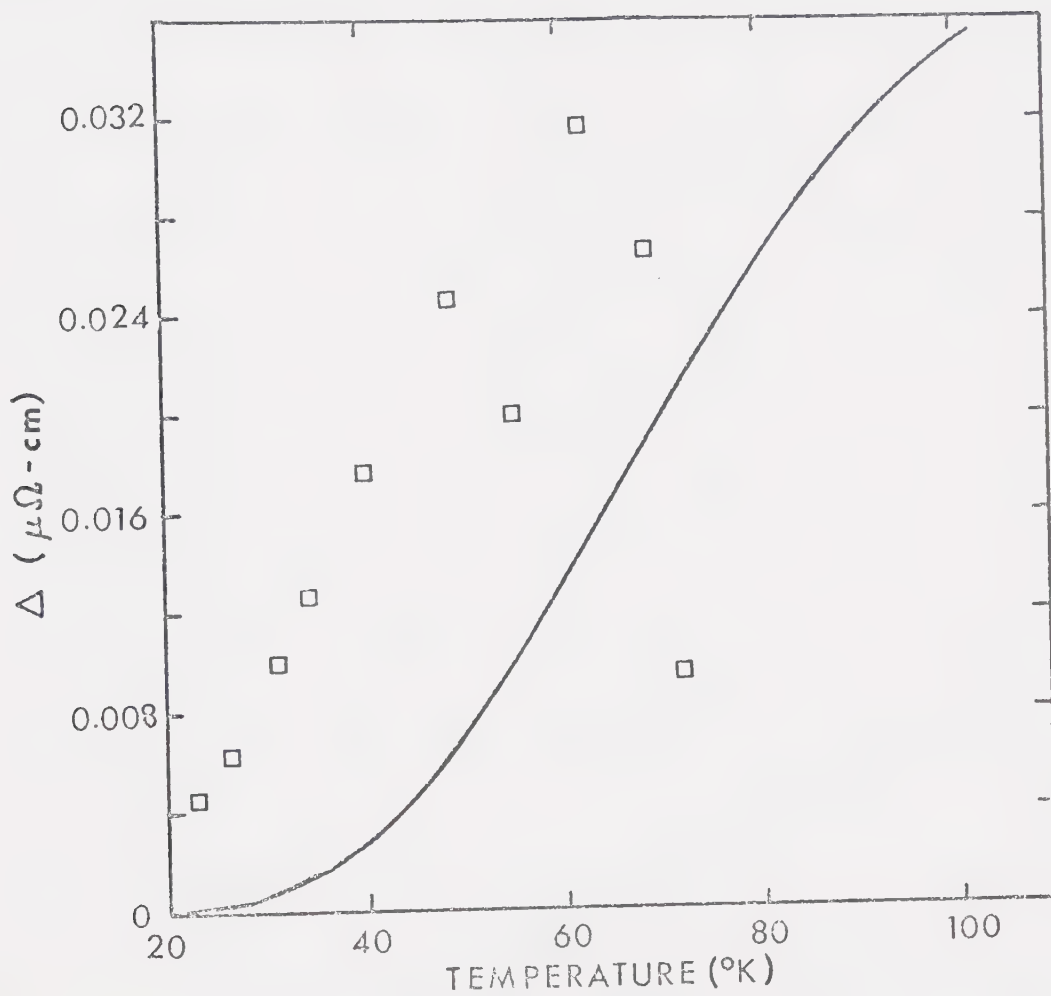


Fig.7

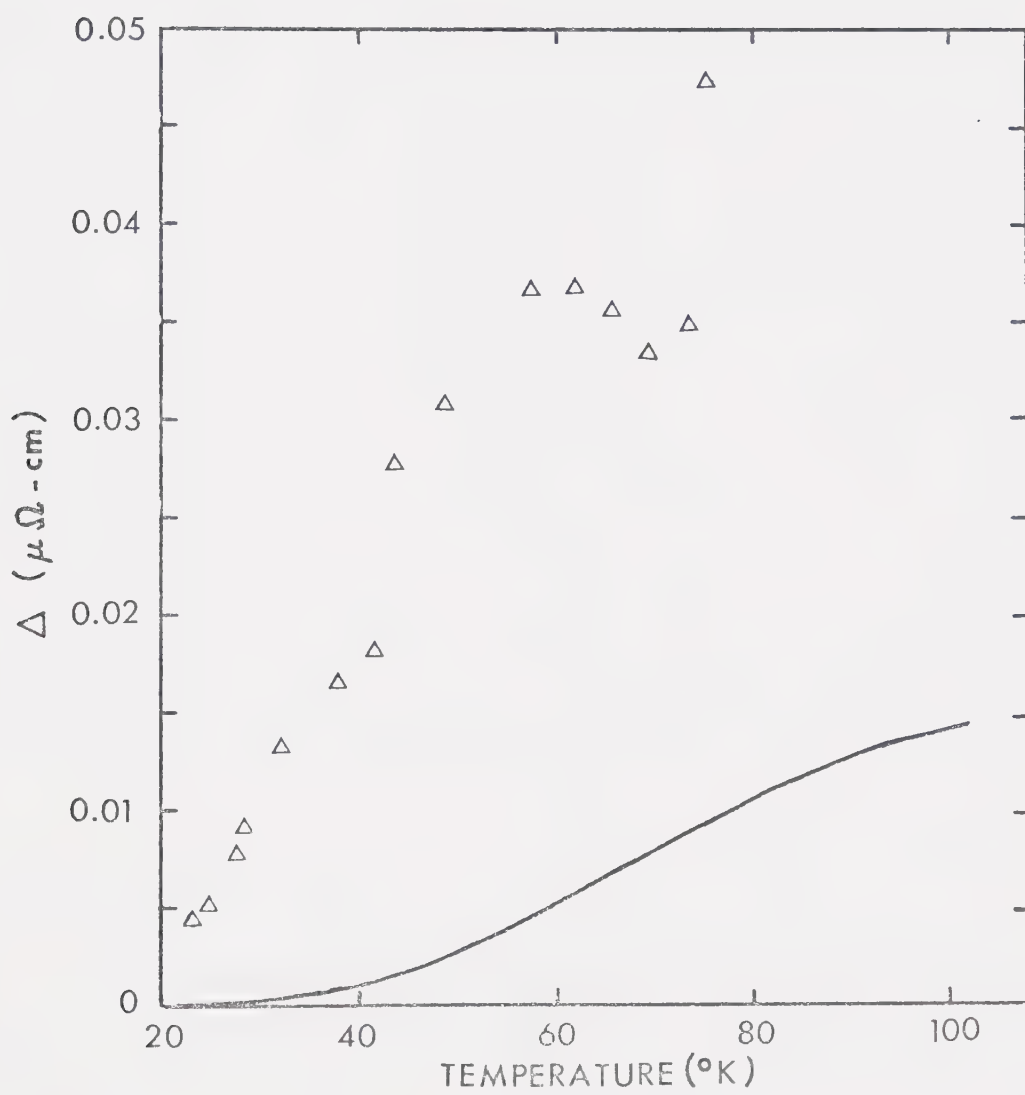


Fig.8

the pure bulk metal. Once ρ_0 is known at different temperatures ℓ can be calculated and hence the resistivity of the film from eqn. (1.9) (Larson, 1971).

In Figs. 6, 7 and 8 the DMR calculated from Fuchs theory and the observed deviations have been plotted. It is seen that the observed deviations are higher than expected from Fuchs theory. This is to be expected as there are other factors such as the presence of impurities and lattice defects in the film and the small angle scattering mechanism (which can be important especially when the scattering at the boundaries is diffuse) which can contribute to the observed deviations. Due to the limited accuracy of the observed deviations and also due to the assumptions made in calculating the deviations expected from Fuchs theory, no rigorous comparison can be made with the theory or with other experimental data.

5. Conclusion

We have obtained electrical resistivity data at room temperature, at 77°K and at 4.2°K on thin aluminum films (50 Å - 10,000 Å) vacuum evaporated at room temperature on glass substrates. We have discussed these data in the light of Fuchs theory and have estimated the value of $\rho_0 \ell$ for these films to be less than $8.2 \times 10^{-12} \Omega \cdot \text{cm}^2$ (at room temperature). We believe the observed

dependence of thin film resistivity on thickness cannot be due to Fuchs size effect alone but must also be due to other thickness dependent factors which contribute to the increase in the resistivity of the films. Unless these factors are eliminated no unambiguous comparison with Fuchs theory can be made.

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